Synthesis and optical characterization of Eu-doped Y$_2$O$_3$ and [(Y,Gd)$_2$O$_3$] phosphor by improved method

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Abstract: Phosphor plays an important role in high-resolution display devices, remote thermometry, gas sensors and thermo mechanic devices. Improved method for the preparation of europium-doped yttrium oxide red phosphor with the inclusion of alkali metal has been reported. Phosphor having the general formula (Y$_{1-x}$Eu$_x$)$_2$O$_3$: MX where 0.025 ≤ x ≤ 0.2 and MX is alkali halide in the mole ratio 0.025 to 0.6, more preferably between 0.05 to 0.2 moles are reported. Another series of red phosphor materials with the general formula (Y$_{1-x-y}$Gd$_x$Euy)$_2$O$_3$: MX where x = 0.1 and 0.025 ≤ y ≤ 0.2 and MX is alkali halide in the mole ratio 0.025 to 0.5 more preferably between 0.1 to 0.2 moles has been reported. Luminescence of phosphor increased with the inclusion of alkali halide. The materials are very soft with fine crystal size less than the 100 nm.

Keywords: phosphor; luminescence; Sharp emission; optical properties.

Introduction
Phosphor can be found in broad range of the every day applications such as cathode ray tubes (CRT) used in medical imaging, projection television (PTVs), fluorescent lights (FL), graphics, electro-luminescent panels (EL) and field emission display (FED) [1-6]. Theses materials provide high luminescence due to the electronic transition mainly due to the 4f → 4f transition of the activator ions.

Disadvantage of CRT for many applications are due to its size and power consumption. However, field emission display solves many problems of the conventional CRT displays. For FED system very small particle size is required instead of larger.

The yttrium oxide phosphor is synthesized by various methods such as thermal, CVD, liquid phase epitaxy, spray pyrolysis and sol-gel methods have been reported [7-9]. All these have their advantage and limitations. In these multi step processes, heating at high temperature usually 1100º C for many hours is required. This made the synthesized material bigger in size.

In this present work the phosphors are synthesized via a new method by using microwave oven. The combustion is done in the microwave. The synthesized phosphors are having luminescence and small size particles.

Experimental
All the chemicals used are of high purity (Aldrich 99.99). The appropriate amounts of yttrium nitrate, europium nitrate and alkali halide (NaCl) are mixed with stoichiometric amount of Y$_2$O$_3$:Eu$^{3+}$,MX and (Y,Gd)$_2$O$_3$:Eu$^{3+}$,MX. In proportionate amount the fuel was also calculated as reported by Ekambaram and Patil [10] and then added to the mixtures. The urea was used as fuel for the combustion reactions.

Then the mixtures were used to propagate for the combustion reactions with in the microwave (Sam-sung RE-CM160S with specification 2450MHz and output 1600 Watts) just for 2-3 minutes. As soon as microwave started, the reaction initiated in oven, flame appears on the surface and spread over the entire volume, leaving a white powder with an extremely porous material. Mixtures were powdered with zirconia balls.

The phosphor materials with general formula (Y$_{1-x}$Eu$_x$)$_2$O$_3$:MX (0.05<x<0.2) and (Y$_{1-y-x}$Gd$_x$Euy)$_2$O$_3$:MX (0.025<y<0.20) were prepared by dissolving a mixtures of chemical in proportionate amount as stated above.

Results and Discussion
Homogenous samples of the europium doped yttrium oxide were prepared by microwave combustion method with in a few minutes with nitrogen based fuel i.e. urea. Though urea is cheap, commercially and easily available fuel so we have used it for the combustion of the metal nitrates. During the combustion the metal nitrates are impregnated into polymeric product and get ignited. A large numbers of gases liberated during the reaction, which made the materials porous, and particles size becomes small.

Inclusion of alkali halides between 0.025 to 0.6 moles with respect to the phosphor material in the above said series with dissolving the metal nitrates greatly increased the intensity of the luminescence of the material. It was noticed that the amount of alkali halides should be between 0.05 to 0.2 moles when luminescence is more than double. Lower value of alkali halides (0.05) gave less luminescence while maximum was reported in the 0.25 moles of alkali halides but material lost softness.

The luminescent materials prepared by the present method are characterized by X-ray diffraction (XRD) spectrum was taken by Rigaku Spectrophotometer model D/max2000 (Japan) with CuKα radiation. Fig. 1 is the XRD patterns obtained for red phosphor composition.
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Figure 1: XRD patterns of
(a) \((Y_{0.95}Eu_{0.05})_2O_3\) without alkali halide
(b) \((Y_{0.95}Eu_{0.05})_2O_3:0.01NaCl\)

Photoluminescence spectra were recorded by using the Minolta CS 1000 series spectrophotometer (USA) and emission was also reported found in red region at 612 nm as shown in the Fig.3 and 4.
Powder morphology of the phosphor was determined by the scanning electron microscope using a Philips XL series max. The particle size was found to be smaller than 100nm and soft also. The SEM of these phosphors is shown in the Fig 5 and 6.
Elemental analyses were also checked by the Philips EDAX PV 9900 series.

Figure 2: is the XRD patterns
(a) \((Y_{0.65}Gd_{0.30}Eu_{0.05})_2O_3\) without alkali halides
(b) \((Y_{0.65}Gd_{0.30}Eu_{0.05})_2O_3:0.01NaCl\)

(a) \((Y_{0.95}Eu_{0.05})_2O_3\) without adding any alkali halide (b) \((Y_{0.95}Eu_{0.05})_2O_3:0.01NaCl\) with alkali halide. Fig. 2 is the XRD patterns obtained for phosphor composition (a) \((Y_{0.65}Gd_{0.30}Eu_{0.05})_2O_3\) without adding any alkali halide (b) \((Y_{0.65}Gd_{0.30}Eu_{0.05})_2O_3:0.01NaCl\) with alkali halide. XRD patterns, which are shown in Fig 1 & 2, reflect that with the inclusion of alkali halides the peaks are found to be sharp which showed the crystalline nature of the phosphors material.

Figure 3: PL spectrum of \((Y_{0.95}Eu_{0.05})_2O_3:0.01NaCl\)
Figure 4: PL spectrum of \((Y_{0.65}Gd_{0.30}Eu_{0.05})_2O_3:0.01NaCl\)

Figure 5: SEM of \((Y_{0.95}Eu_{0.05})_2O_3:0.01NaCl\)
Figure 6: SEM \((Y_{0.65}Gd_{0.30}Eu_{0.05})_2O_3:0.01NaCl\)
Conclusion
Nano scale \( \text{Y}_2\text{O}_3;\text{Eu}^{3+},\text{MX} \) and \((\text{Y},\text{Gd})_2\text{O}_3;\text{Eu}^{3+},\text{MX}\) have been prepared by the facile microwave combustion syntheses. The microwave synthesis is fast, less time consuming, energy saver process, which produces fine, chemically homogenous, pure and simple phase phosphor material. However luminescence is also greatly increased in this series of phosphors by the addition of alkali halides.

References